

PATENT COOPERATION TREATY

PCT

INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

(Chapter II of the Patent Cooperation Treaty)

(PCT Article 36 and Rule 70)

REC'D 03 FEB 2006

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Applicant's or agent's file reference BPCL 10002	FOR FURTHER ACTION		See Form PCT/APEA/416
International application No. PCT/GB2005/000438	International filing date (day/month/year) 09.02.2005	Priority date (day/month/year) 03.03.2004	
International Patent Classification (IPC) or national classification and IPC C07C51/12, C07C67/36, C07C53/08, C07C69/14			
Applicant BP CHEMICALS LIMITED et al.			
<p>1. This report is the international preliminary examination report, established by this International Preliminary Examining Authority under Article 35 and transmitted to the applicant according to Article 36.</p> <p>2. This REPORT consists of a total of 6 sheets, including this cover sheet.</p> <p>3. This report is also accompanied by ANNEXES, comprising:</p> <p>a. <input type="checkbox"/> <i>sent to the applicant and to the International Bureau</i> a total of sheets, as follows:</p> <ul style="list-style-type: none"> <input type="checkbox"/> sheets of the description, claims and/or drawings which have been amended and are the basis of this report and/or sheets containing rectifications authorized by this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions). <input type="checkbox"/> sheets which supersede earlier sheets, but which this Authority considers contain an amendment that goes beyond the disclosure in the international application as filed, as indicated in item 4 of Box No. I and the Supplemental Box. <p>b. <input type="checkbox"/> <i>(sent to the International Bureau only)</i> a total of (indicate type and number of electronic carrier(s)), containing a sequence listing and/or tables related thereto, in computer readable form only, as indicated in the Supplemental Box Relating to Sequence Listing (see Section 802 of the Administrative Instructions).</p>			
<p>4. This report contains indications relating to the following items:</p> <ul style="list-style-type: none"> <input checked="" type="checkbox"/> Box No. I Basis of the opinion <input type="checkbox"/> Box No. II Priority <input type="checkbox"/> Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability <input type="checkbox"/> Box No. IV Lack of unity of invention <input checked="" type="checkbox"/> Box No. V Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement <input type="checkbox"/> Box No. VI Certain documents cited <input type="checkbox"/> Box No. VII Certain defects in the international application <input checked="" type="checkbox"/> Box No. VIII Certain observations on the international application 			
Date of submission of the demand 29.09.2005	Date of completion of this report 01.02.2006		
Name and mailing address of the international preliminary examining authority:  European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized Officer Molina de Alba, J Telephone No. +49 89 2399- 		

INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

International application No.
PCT/GB2005/000438

Box No. I Basis of the report

1. With regard to the **language**, this report is based on the international application in the language in which it was filed, unless otherwise indicated under this item.
 - This report is based on translations from the original language into the following language , which is the language of a translation furnished for the purposes of:
 - international search (under Rules 12.3 and 23.1(b))
 - publication of the international application (under Rule 12.4)
 - international preliminary examination (under Rules 55.2 and/or 55.3)
 2. With regard to the **elements*** of the international application, this report is based on (*replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report*):

Description, Pages

1-9 as originally filed

Claims, Numbers

1-33 as originally filed

- a sequence listing and/or any related table(s) - see Supplemental Box Relating to Sequence Listing

3. The amendments have resulted in the cancellation of:

 - the description, pages
 - the claims, Nos.
 - the drawings, sheets/figs
 - the sequence listing (*specify*):
 - any table(s) related to sequence listing (*specify*):

4. This report has been established as if (some of) the amendments annexed to this report and listed below had not been made, since they have been considered to go beyond the disclosure as filed, as indicated in the Supplemental Box (Rule 70.2(c)).

 - the description, pages
 - the claims, Nos.
 - the drawings, sheets/figs
 - the sequence listing (*specify*):
 - any table(s) related to sequence listing (*specify*):

* If item 4 applies, some or all of these sheets may be marked "superseded."

INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

International application No.
PCT/GB2005/000438

Box No. V Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes:	Claims	1-33
	No:	Claims	
Inventive step (IS)	Yes:	Claims	
	No:	Claims	1-33
Industrial applicability (IA)	Yes:	Claims	1-33
	No:	Claims	

2. Citations and explanations (Rule 70.7):

see separate sheet

Box No. VIII Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

see separate sheet

**INTERNATIONAL PRELIMINARY
REPORT ON PATENTABILITY
(SEPARATE SHEET)**

International application No.
PCT/GB2005/000438

1) Reference is made to the following documents:

- D1:** EP-A-0 353 722 (UNION CARBIDE CHEMICALS AND PLASTICS COMPANY INC.) 7 February 1990 (1990-02-07)
- D2:** WO 98/57918 A (BP CHEMICALS LIMITED; JONES, MICHAEL, DAVID) 23 December 1998 (1998-12-23)
- D3:** WO 03/014054 A (EASTMAN CHEMICAL COMPANY) 20 February 2003 (2003-02-20)
- D4:** WO 01/07393 A (UNION CARBIDE CHEMICALS & PLASTICS) 1 February 2001 (2001-02-01)

2) The present application relates to a carbonylation process for the production of a carbonylation product by contacting carbon monoxide with a feed comprising an alcohol and/or a reactive derivative thereof in the vapour phase using an heterogeneous heteropolyacid catalyst comprising one or more metal cations selected from Cu, Fe, Ru, Os, Co, Rh, Ir, Ni, Pd, and Pt, characterized in that there is also present in the feed at least 0.5 wt.% water.

3) Re Item V

3.1 Novelty (Art. 33(2) PCT)

D1 discloses (cf. abstract and pg. 3, l. 39-53) a process for the carbonylation of one or more alcohols and ethers to esters and/or carboxylic acids. The reaction is effected in the vapour state over a heterogeneous heteropolyacid catalyst which is preferably deposited on a support inert to the reaction such as silica. The use of a heteropolyacid catalyst circumvents the use of methyl iodide as a promoter, which in the vapour phase is corrosive and requires expensive material (cf. pg. 3, l. 3-6). The most preferred temperature of the process is between 150°C and 300°C and the pressure from 1 to 35 atmosphere (cf. pg. 5, l. 39-57). The examples illustrate such a process, wherein methanol or dimethyl ether are converted to methyl acetate and/or acetic acid at 225°C and 1 atm with a methyl acetate selectivity greater than 95% (cf. pg. 7, l. 15-17 and examples 1-3, 5-7, 11-18, and 24-33). The subject-matter of the present application differs from **D1** in that it contains at least 0.5 wt.% in the feed.

**INTERNATIONAL PRELIMINARY
REPORT ON PATENTABILITY
(SEPARATE SHEET)**

International application No.
PCT/GB2005/000438

D2 relates to (cf. abstract) a process for the carbonylation of an alcohol or a reacting derivative thereof in the liquid phase at elevated temperature and pressure in the presence of a heterogeneous catalyst supported on a polymeric resin. The process requires a halogen promoter and water as well as the presence of hydrogen in the carbon monoxide feed. Water and hydrogen are essential components. Water is present in an amount between 0.1 wt.% and up to 25 wt.% and it accelerates the carbonylation reaction and improves the selectivity to the desired product (cf. pg. 1, l. 8-11 and pg. 4, l. 20-30). Hydrogen, present in 0.1 to 10 bar, reduces leaching of the catalyst from the support in the presence of water (cf. pg. 2, l. 33-pg. 3, l. 3 and pg. 4, l. 34-pg. 35, l. 5). The presently claimed process differs from the process of **D2** in that the heterogeneous catalyst is a heteropolyacid and in that the process is carried out in the vapour phase.

D3 also describes (cf. abstract and pg. 8, l. 8-15) a carbonylation process of lower aliphatic alcohols or ethers for producing carboxylic acids and/or esters. The process is performed in vapour phase in the presence of a heterogeneous catalyst supported on a solid material. Although the presence of water in the gaseous feed is not essential when carbonylating methanol, it serves to suppress formation of methyl acetate and/or dimethyl ether. When using an alternative source of methanol such as dimethyl ether, the amount of water fed used usually is increased to account for the mole of water required for hydrolysis of the methanol alternative. The addition of water has thus a primary utility for the preparation of acetic acid (cf. pg. 9, l. 15-31). The process of Claim 1 differs from that of **D2** in that the heterogeneous catalyst is a heteropolyacid.

As none of **D1-D3** discloses a carbonylation process as presently claimed, the subject-matter of claims 1-33 is regarded as novel.

3.2 Inventive Step (Art. 33(3) PCT)

D1 is considered to represent the closest state of the art. As already mentioned, the claimed process differs from **D1** in that it contains at least 0.5 wt.% water in the feed. According to the examples filed within the application, the addition of water to a process a described in **D1** results in an improvement of the catalyst activity (enhanced product selectivity). The problem to be solved may thus be regarded as the provision of a carbonylation process which has a higher product selectivity.

**INTERNATIONAL PRELIMINARY
REPORT ON PATENTABILITY
(SEPARATE SHEET)**

International application No.
PCT/GB2005/000438

The solution provided by the present application to the problem posed, namely the addition of water to the feed, would be obvious to the skilled person in the light of the documents **D2** and **D3**, which indicate an enhancement in the product selectivity among other positive properties derived from the presence of water in the process. An inventive step may thus not be acknowledged for the present application.

3.3 Industrial applicability (Art. 33(4) PCT)

Is acknowledged for claims 1-33.

4) Re Item VIII

4.1 Claim 1 is not clear (Art. 6 PCT) for two reasons:

- I. The claim relates to a carbonylation process in the **vapour phase**. However, in the illustrating examples, it seems that methanol and water are added and reacted in liquid phase (cf. pg. 7, l. 17-20). This inconsistency renders the scope of Claim 1 unclear.
- ii. The feature "reactive derivative thereof" is too broad (Art. 6 and 33(3) PCT) and it is not clear which derivatives should be understood as encompassed by such a formulation. There are derivatives, e.g. olefines, with would react in a different manner as alcohols do, and for which the advantages shown in the application may not be extrapolated. Other derivatives are simply not suitable to undergo a carbonylation reaction.

4.2 Claim 2 is inconsistent in that it relates to a process according to Claim 1 wherein the feed **comprises 1 wt.% water** but mentions afterwards "such as **at least 2 wt.%,** preferably **at least 5 wt.%**".

4.3 The word "acid" lacks in Claim 24 after the first "carboxylic".